

REMARKS

This second preliminary amendment is submitted prior to examination of the above-identified Divisional Patent Application.

The amendments to the specification at pages 11-13, 38, 41, 42 and 44 correct obvious errors, as the drawing Figures in question contain parts a and b, and in the case of Figure 18, contain parts a, b and c. These parts are shown in the actual drawings. Thus no new matter is introduced.

In the parent application the number "20" was inadvertently skipped in the claim numbering sequence. In the parent application, the Examiner required applicants to submit a renumbered claim set. In this second preliminary amendment applicants have renumbered claims 45-57 and 61-69 as 44-56 and 60-68. Originally numbered claims 58-60 and 70 from the application were cancelled without prejudice in first preliminary amendment. In this second preliminary amendment these claims have been rewritten without multiple dependencies as claims 71-88. This does not introduce new matter. All of the claims are amended to conform to United States practice, as well as to the subject matter allowed in the parent application.

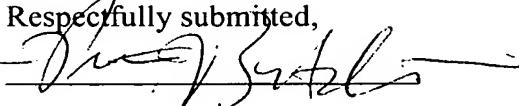
Attached hereto is a marked-up version of the changes made to the specification and claims. The attached page is captioned "Version with markings to show changes made." A favorable first action on the merits is respectfully requested. The Examiner is requested to telephone the undersigned if there are any issues requiring resolution.

Finally, the Examiner is authorized to charge Applicant's Deposit Account No. 19-5425 for any charges in connection with this Preliminary Amendment.

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Respectfully submitted,


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VERSION WITH MARKINGS TO SHOW CHANGES MADE

In the specification:

The three paragraphs at page 11, lines 12-24 have been amended as follow:

~~Figure 8 shows Figures 8a and 8b show~~ the second cycle voltage verses capacity plot for the battery containing M30 activated carbon pyrolyzed at 1000°C of the Illustrative Example re activated carbons.

~~Figure 9 shows Figures 9a and 9b show~~ the first cycle voltage versus capacity plot for the battery containing M30 activated carbon pyrolyzed at 1000°C of the Illustrative Example re activated carbons.

~~Figure 10 compares Figures 10a and 10b compare~~ the second cycle voltage versus capacity plots of sample no. I of Epoxy Example 1 to that of the 700°C pyrolyzed sample of Prior Art Example 2.

The paragraph at page 12, lines 24-26 has been amended as follows:

~~Figure 16 shows Figures 16a and 16b show~~ the voltage versus capacity plot of the first discharge and charge of the battery comprising sample no. VII of Epoxy Example 1.

The paragraph from page 12, line 33 to page 13, line 2 has been amended as follows:

~~Figures 18a ,and 18b and 18c~~ show the voltage versus capacity plots for the first and second cycles respectively for batteries comprising samples prepared from the A type precursor in Phenolic Resin Example 1. The curves have been offset sequentially for clarity. (In ~~both~~ the Figures, the shifts are 0.0, 0.15, 0.3, 0.45; and 0.7 volts for ~~sample~~ samples A700, A800, A900, A1000; and A1100 respectively.)

The paragraph at page 13, lines 27-30, has been amended as follows:

~~Figure 22 shows Figures 22a and 22b show~~ the voltage versus capacity plots for the second cycle of batteries comprising samples prepared from the B type precursor in Phenolic Resin Example 2. The plots have been sequentially offset by 0.1V for clarity.

The paragraph at page 38, lines 4-15, has been amended as follows:

All samples exhibited high specific capacities but also substantial hysteresis in the voltage plot and substantial irreversible capacity on the first discharge. For example, ~~Figure 8 shows Figures 8a and 8b show~~ the second cycle voltage versus capacity plot for the battery containing M30 activated carbon pyrolyzed at 1000°C. The specific capacity is about 550 mAh/g and there is substantial hysteresis. ~~Figure 9 shows Figures 9a and 9b show~~ the first cycle voltage verses capacity plot for the same battery containing M30 activated carbon pyrolyzed at 1000°C. The first discharge capacity is enormous at about 2000 mAh/g and thus there is substantial irreversible capacity.

The paragraph at page 41, lines 1-16, has been amended as follows:

The voltage versus capacity plots for sample no. I pyrolyzed at 700°C is compared to that of the pitch sample of Prior Art Example 2 pyrolyzed at the same temperature in ~~Figure 10 Figures 10a and 10b~~. These two plots show almost identical behaviour (although the battery using sample no. I was allowed to plate more lithium). Figure 4 indicates that the two samples in ~~Figure 10 Figures 10a and 10b~~ have almost the same H/C ratio. Figure 11 shows the x-ray diffraction patterns of samples no. I, II, and III (offset by 1600 counts). Therein, it can be seen that sample no. I has a substantially smaller R than the corresponding pitch sample in Figure 5. There are very few stacked graphene layers in sample no. I as evidenced by the {002} peak amounting to only a shoulder on the low angle background. Figures 11 and 5 also show that these structural differences persist at higher pyrolysis temperatures.

The paragraph at page 42, lines 7-20, has been amended as follows:

~~Figure 16 shows Figures 16a and 16b show~~ the first discharge and charge of the laboratory coin cell battery employing sample no. VII. The battery shows a first discharge capacity of about

625 mAh/g and the first recharge capacity of about 465 mAh/g. The irreversible capacity of sample VII is therefore only about 160 mAh/g, which is considered to be in an acceptable range for practical lithium ion batteries. The surface area measured by the conventional BET method for sample VII was 217 m²/g. If this area were all accessible to electrolyte, such low values for the irreversible capacity would not be expected (for example, based on the disclosure of U.S. Patent No. 5,028,500). However, the MB adsorption capacity is relatively low (<5 micromoles/g) for this and the other invention samples tested.

The paragraph at page 46, line 1-15, has been amended as follows:

~~Figure Figures 18a and 18b show shows~~ the first discharge-charge cycle for the series of pyrolyzed A type precursors. The samples heated at 700°C and 800°C show significant hysteresis in the voltage profile (Li is inserted near 0V but is removed near 1.0V). This has been ascribed to the large hydrogen content in the samples. Upon heated to 900°C, the hysteresis is predominantly eliminated and the samples show substantial capacity at low voltage. Figure 18b ~~18c~~ shows the second cycle of the same series. The vertical lines indicate the onset of lithium plating during discharge. The batteries prepared from material heated to 900°C and 1000°C appear most promising for this series. Their reversible capacities are about 510 and 450 mAh/g respectively.

In the claims:

Claims 45-57 and 61-69 have been amended as follows:

~~45. 44.~~ A process for preparing a pre-graphitic carbonaceous host for a carbonaceous insertion compound comprising pyrolyzing an epoxy precursor ~~at a temperature above 700°C,~~ or a phenolic resin precursor ~~at a temperature above 800°C,~~ or a carbohydrate precursor or a carbohydrate containing precursor at a temperature above 800°C, and within a temperature range effective to produce an H/C atomic ratio less than about 0.1 and an ~~such that the~~ empirical parameter R for said host wherein R:

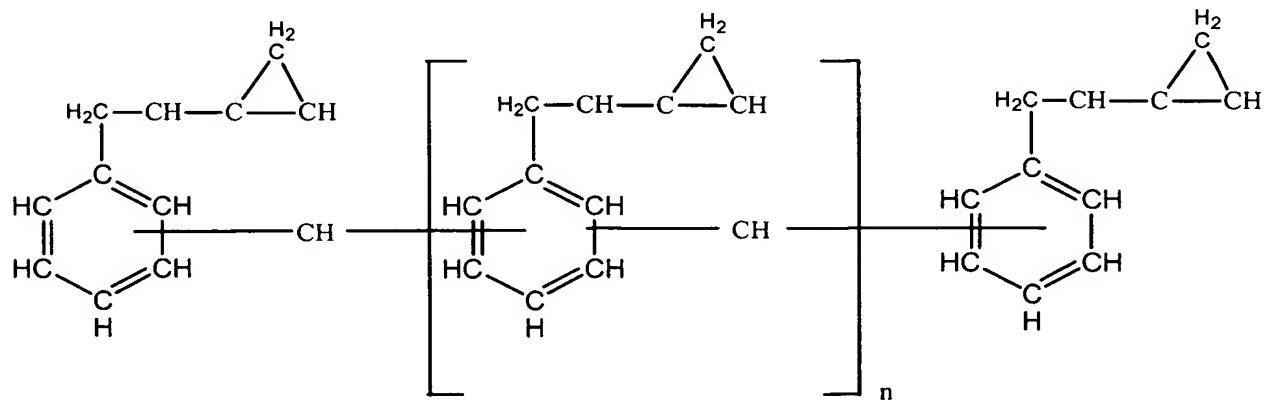
(i) is determined by X-ray from an x-ray diffraction pattern using a diffractometer equipped with a copper target X-ray tube and a diffracted beam monochromometer, with the X-ray beam of said diffractometer being confined to the sample in the angular range from 10° to 35° in scattering angle,

(ii) and is defined as the {002} peak divided by the background level, and

(iii) is less than about 2.2.

~~46. 45. A process as claimed in claim 44, for preparing a pre-graphitic carbonaceous host for a carbonaceous insertion compound comprising pyrolyzing an epoxy precursor at a temperature above 700°C such that the empirical parameter R, determined from an x-ray diffraction pattern and defined as the {002} peak divided by the background level, is less than about 2.2.~~

~~47. 46. A process as claimed in claim 46 45 wherein the epoxy precursor is an epoxy novolac resin with formula~~

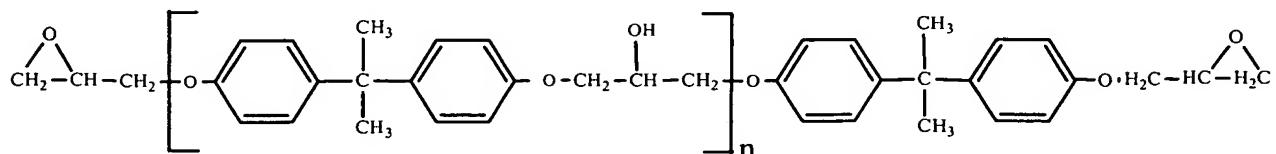


Epoxy Novolac Resin

$n = 1.6$

and the pyrolysis is performed at a maximum temperature below about 1100°C.

48. 47. A process as claimed in claim 46 45 wherein the epoxy precursor is a bisphenol A epoxy resin with formula



Bisphenol-A Epoxy Resin

n = 12

and the pyrolysis is performed at a temperature about 800°C.

49. 48. A process as claimed in claim 44. for preparing a pre-graphitic carbonaceous host for a carbonaceous insertion compound comprising pyrolyzing a phenolic resin precursor at a temperature above 800°C such that the empirical parameter R, determined from an x-ray diffraction pattern and defined as the {002} peak divided by the background level, is less than about 2.2.

50. 49. A process as claimed in claim 49 48 wherein the phenolic resin precursor is of the novolac type.

51. 50. A process as claimed in claim 49 48 wherein the phenolic resin precursor is of the resole type.

52. 51. A process as claimed in claim 51 50 wherein the pyrolysis is performed at a temperature in the range from about 900°C to about 1100°C.

53. 52. A process as claimed in claim 44, for preparing a pre-graphitic carbonaceous host for a carbonaceous insertion compound comprising pyrolyzing a carbohydrate precursor or a carbohydrate containing precursor at a temperature above 800°C such that the empirical parameter R, determined from an x-ray diffraction pattern and defined as the {002} peak divided by the background level, is less than about 2.2.

54. 53. A process as claimed in claim 53 52 wherein the carbohydrate precursor is selected from the group consisting of sugar, starch, and cellulose.

55. 54. A process as claimed in claim 53 52 additionally comprising precarbonizing the carbohydrate by washing with an acid.

56. 55. A process as claimed in claim 55 54 wherein the carbohydrate is sucrose.

57. 56. A process as claimed in claim 55 54 wherein the acid is concentrated sulfuric acid.

61. 60. An electorchemical device comprising The use of a carbonaceous insertion compound in an electrode, wherein at least a portion of said electrode comprises a carbonaceous insertion compound of an electrochemical device, said carbonaceous insertion compound comprising: a pre-graphitic carbonaceous host prepared by the method of claim 44 pyrolyzing an epoxy precursor at a temperature above 700°C, or a phenolic resin precursor at a temperature above 800°C, or a carbohydrate precursor, or a carbohydrate containing precursor, at a temperature above 800°C, wherein the empirical parameter R, determined from an x-ray diffraction pattern and defined as the {002} peak height divided by the background level, is less than about 2.2; and atoms of an alkali metal inserted into the carbonaceous host.

62. 61. An electrochemical device as claimed in claim 60, wherein ~~The use of a carbonaceous insertion compound in an electrode of an electrochemical device, said carbonaceous insertion compound comprising:~~ comprises a pre-graphitic carbonaceous host prepared by pyrolyzing an epoxy precursor at a temperature above 700°C wherein the empirical parameter R, determined from an x-ray diffraction pattern and defined as the {002} peak height divided by the background level, is less than about 2.2, and atoms of an alkali metal inserted into the carbonaceous host.

63. 62. An electrochemical device ~~The use of the carbonaceous insertion compound as claimed in claim 62~~ 61 wherein the epoxy precursor is a novolac epoxy resin.

64. 63. An electrochemical device ~~The use of the carbonaceous insertion compound as claimed in claim 62~~ 61 wherein the epoxy precursor is a bisphenol A epoxy resin.

65. 64. An electrochemical device as claimed in claim 60, wherein ~~The use of a carbonaceous insertion compound in an electrode of an electrochemical device, said carbonaceous insertion compound comprising:~~ comprises a pre-graphitic carbonaceous host prepared by pyrolyzing a phenolic resin precursor at a temperature above 800°C wherein the empirical parameter R, determined from an x-ray diffraction pattern and defined as the {002} peak height divided by the background level, is less than about 2.2, and atoms of an alkali metal inserted into the carbonaceous host.

66. 65. An electrochemical device ~~The use of the carbonaceous insertion compound as claimed in claim 65~~ 64 wherein the phenolic resin precursor is of the novolac type.

67. 66. An electrochemical device ~~The use of the carbonaceous insertion compound as claimed in claim 65~~ 64 wherein the phenolic resin precursor is of the resole type.

68. 67. An electrochemical device as claimed in claim 60, wherein ~~The use of a carbonaceous insertion compound in an electrode of an electrochemical device, said carbonaceous insertion compound comprising:~~ comprises a pre-graphitic carbonaceous host prepared by pyrolyzing a carbohydrate precursor; or a carbohydrate containing precursor, at a temperature above 800°C wherein the empirical parameter R, determined from an x-ray diffraction pattern and defined as the {002} peak height divided by the background level, is less than about 2.2, and atoms of an alkali metal inserted into the carbonaceous host.

69. 68. An electrochemical device ~~The use of the carbonaceous insertion compound as claimed in claim 68~~ 67 wherein the carbohydrate precursor is selected from the group consisting of sugar, starch, and cellulose.

Claims 71-88 have been added:

71. An electrochemical device comprising an electrode wherein a portion of the electrode comprises a carbonaceous insertion compound comprising:

(a) a pre-graphitic carbonaceous host having a reversible capacity for lithium insertion, an irreversible capacity for lithium insertion, and a surface area accessible to a non-aqueous electrolyte wherein

(i) the empirical parameter R for said carbonaceous insertion compound:

(A) is, determined by X-ray diffraction using a diffractometer equipped with a copper target X-ray tube and a diffracted beam monochromator, with the X-ray beam of said diffractometer being confined to the sample in the angular range from 10° to 35° in scattering angle,

(B) is defined as the height of the centre of the {002} peak divided by the background level, and

(C) is less than about 2.2;

(ii) the H/C atomic ratio is less than about 0.1; and

(iii) the electrolyte accessible surface area is sufficiently small such that the irreversible capacity is less than about a half that of the reversible capacity;

(b) and alkali metal atoms reversibly inserted into the carbonaceous host in an amount greater than that which can be reversibly inserted into graphite.

72. An electrochemical device according to claim 71, wherein the alkali metal is lithium.

73. A battery comprising an electrode wherein a portion of the electrode comprises a carbonaceous insertion compound comprising:

(a) a pre-graphitic carbonaceous host having a reversible capacity for lithium insertion, an irreversible capacity for lithium insertion, and a surface area accessible to a non-aqueous electrolyte wherein

(i) the empirical parameter R for said carbonaceous insertion compound:

(A) is, determined by X-ray diffraction using a diffractometer equipped with a copper target X-ray tube and a diffracted beam monochromometer, with the X-ray beam of said diffractometer being confined to the sample in the angular range from 10° to 35° in scattering angle,

(B) is defined as the height of the centre of the {002} peak divided by the background level, and

(C) is less than about 2.2;

(ii) the H/C atomic ratio is less than about 0.1; and

(iii) the electrolyte accessible surface area is sufficiently small such that the irreversible capacity is less than about a half that of the reversible capacity;

(b) and alkali metal atoms reversibly inserted into the carbonaceous host in an amount greater than that which can be reversibly inserted into graphite.

74. A battery according to claim 73, wherein the alkali metal is lithium.

75. A battery comprising an electrode wherein a portion of the electrode comprises a carbonaceous insertion compound prepared according to the process of claim 44.

76. A battery as claimed in claim 75, wherein a portion of the electrode comprises a carbonaceous insertion compound is prepared by pyrolyzing an epoxy precursor compound comprising an epoxy novolac resin.

77. A battery as claimed in claim 75, wherein a portion of the electrode comprises a carbonaceous insertion compound prepared by pyrolyzing an epoxy precursor compound comprising a Bisphenol A epoxy resin.

78. A battery comprising an electrode wherein a portion of the electrode comprises a carbonaceous insertion compound prepared by the process of claim 48.

79. A battery comprising an electrode wherein a portion of the electrode comprises a carbonaceous insertion compound prepared by the process of claim 52.

80. A non-aqueous battery comprising: a cathode comprising a lithium insertion compound; a non-aqueous electrolyte comprising a lithium salt dissolved in a mixture of non-aqueous solvents; and an anode comprising a carbonaceous insertion compound comprising:

(a) a pre-graphitic carbonaceous host having a reversible capacity for lithium insertion, an irreversible capacity for lithium insertion, and a surface area accessible to a non-aqueous electrolyte wherein

(i) the empirical parameter R for said carbonaceous insertion compound: (A) is, determined by X-ray diffraction using a diffractometer equipped with a copper target X-ray tube and a diffracted beam monochromator, with the X-ray beam of said diffractometer being confined to the sample in the angular range from 10° to 35° in scattering angle,

(B) is defined as the height of the centre of the {002} peak divided by the background level, and

(C) is less than about 2.2;

- (ii) the H/C atomic ratio is less than about 0.1; and
- (iii) the electrolyte accessible surface area is sufficiently small such that the irreversible capacity is less than about a half that of the reversible capacity;

(b) and lithium atoms reversibly inserted into the carbonaceous host in an amount greater than that which can be reversibly inserted into graphite.

81. A non-aqueous battery comprising: a cathode comprising a lithium insertion compound; a non-aqueous electrolyte comprising a lithium salt dissolved in a mixture of non-aqueous solvents; and an anode comprising a carbonaceous insertion compound prepared by the process of claim 44, wherein the alkali metal is Li.

82. A non-aqueous battery as claimed in claim 81 comprising: a cathode comprising a lithium insertion compound; a non-aqueous electrolyte comprising a lithium salt dissolved in a mixture of non-aqueous solvents; and an anode comprising a carbonaceous insertion compound prepared by pyrolyzing an epoxy precursor compound comprising a novolac resin.

83. A non-aqueous battery as claimed in claim 81 comprising: a cathode comprising a lithium insertion compound; a non-aqueous electrolyte comprising a lithium salt dissolved in a mixture of non-aqueous solvents; and an anode comprising a carbonaceous insertion compound prepared by pyrolyzin an epoxy precursor compound comprising an epoxy novolac resin.

84. A non-aqueous battery comprising: a cathode comprising a lithium insertion compound; a non-aqueous electrolyte comprising a lithium salt dissolved in a mixture of non-aqueous solvents; and an anode comprising a carbonaceous insertion compound prepared by the process of claim 48.

85. A non-aqueous battery comprising: a cathode comprising a lithium insertion compound; a non-aqueous electrolyte comprising a lithium salt dissolved in a mixture of non-aqueous solvents; and an anode comprising a carbonaceous insertion compound prepared by the process of claim 52.

86. The electrochemical device as claimed in claim 62 wherein the alkali metal is lithium and the electrochemical device is a non-aqueous battery, the battery comprising a cathode comprising a lithium insertion compound; a non-aqueous battery electrolyte comprising a lithium salt dissolved in a mixture of non-aqueous solvents; and an anode comprising said carbonaceous insertion compound.

87. The electrochemical device as claimed in claim 65 wherein the alkali metal is lithium and the electrochemical device is a non-aqueous battery, the battery comprising a cathode comprising a lithium insertion compound; a non-aqueous battery electrolyte comprising a lithium salt dissolved in a mixture of non-aqueous solvents; and an anode comprising said carbonaceous insertion compound.

88. The electrochemical device as claimed in claim 68 wherein the alkali metal is lithium and the electrochemical device is a non-aqueous battery, the battery comprising a cathode comprising a lithium insertion compound; a non-aqueous battery electrolyte comprising a lithium salt dissolved in a mixture of non-aqueous solvents; and an anode comprising said carbonaceous insertion compound.